

Joint European Magnetic Symposia

3<sup>rd</sup> – 7<sup>th</sup> September 2018 • Mainz • Germany

General Chalr: Prof. Dr. Jairo Sinova Co-General Chalr: Prof. Dr. Mathias Kläui



# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 1

SP3 - Parallel Session 1

SP3.1.01

### Artificial Spin Ice Inspired Computation using Nanomagnets

H. Arava, P. Derlet, J. Vijayakumar, J. Cui, A. Kleibert, L. Heyderman

**Text** Computation using nanomagnets has the advantage of being a low power solution to current CMOS based technologies and is interesting for "beyond CMOS" applications. Here, we present an alternative design to perform Boolean logic using nanoscale magnets. Within this new design, we introduce nanomagnets in a loop scheme as building blocks, which can be considered as a zero dimensional implementation of the artificial square ice. For any computational technology to be complete, it would need two basic elements: (i) a channel to transport information and (ii) a logic gate to perform computation. Here, a 1D representation of the square ice, a square loop chain, was implemented to serve as a "channel" to transport information and logic gates based on the loop structures were designed based on the energy levels associated with different magnetic vertex charges in an artificial square ice. Each logic operation corresponds to a truth table (such as NAND), and is a consequence of the minimization of dipolar energy associated with the magnetic moment rearrangements. We employed synchrotron X-ray PEEM (Photo-Emission Electron Microscopy) to verify the logic operations that were driven by a thermal protocol. We were successful in obtaining an output from a defined input across a maximum tested chain length comprising of 19 square rings. In addition, we tested over 2000 logic gates and experimentally observed that ~94% displayed correct gate operation.



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SP3 - Parallel Session 1

SP3.1.02

# Study of the features of magnetic ordering in honeycomb layered oxides of transition metals

A. Korshunov, A. Kurbakov, S. Podchezertsev

**Text** Low dimensional magnetism is one of the most fascinating and actively developing topics of modern solid-state physics. Quantum essence of matter manifests itself most vividly in reduced dimension that gives a rise to plenty of new phenomena. The current work is aimed to study the features of long-range spin ordering at low temperature in two dimensional honeycomb oxides. The studied objects represent two groups with structural formulas  $A_2M_2TeO_6$  (monoclinic C2/m) and  $A_3M_2SbO_6$  (hexagonal P6<sub>3</sub>/mcm), where  $A^+ = Li$ , Na and  $M^{2+} = Ni$ , Co. Despite of space groups differences, both families have similar crystal structure: honeycomb layers of magnetic atoms is divided by nonmagnetic alkali atoms. The long-range spin ordering in such lattice is a result of complex competition of AFM and FM exchange interactions up to third magnetic neighbor (so-called  $J_1$ - $J_2$ - $J_3$  model). Our neutron powder diffraction (NPD) studies have shown that all compounds have zigzag spin structure in honeycomb layers with special features that is depended on the internal structure and composition. It should be noted that Ni (3d<sup>8</sup>) and Co (3d<sup>7</sup>) are in high spin state S = 1 and S = 3/2 respectively that directly affects magnetic properties of connected compounds, such as strong anisotropy of magnetic moment direction.

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SP3 - Parallel Session 1

SP3.1.04

### Balanced nanomagnetic logic gate in Kagome spin ice

P. Gypens, J. Leliaert, B. Van Waeyenberge

**Text** Nanomagnetic logic (NML) is a promising candidate to replace or complement traditional chargedbased logic devices. Single NML gates are well studied and their functionality has been verified experimentally. However, such gates suffer from a problem in that they sometimes produce erroneous output when integrated into circuits, due to the fact that the ground state of connected gates can differ from the one in each individual gate. A fundamental solution is offered by using balanced logic gates: gates for which the ground states corresponding to all possible input states have the same energy.

In this contribution, we present a balanced NAND gate consisting of Kagome spin ice elements [1]. In a Kagome spin ice, magnetic nano islands are placed along the edges of a hexagonal lattice, such that three nano islands meet at each vertex. In a ground state configuration, each vertex contains at least one spin pointing inwards and outwards, while the remaining spin can point in either direction. We exploit this fact for the design of a balanced magnetic logic gate. The presented gate provides the first proof-of-concept of an artificial Kagome spin ice NML gate and can be driven either by an out-of-plane clocking field or can be thermally driven. It is shown that the presented gate has a reliability of at least 96%, and is robust against disorder.

[1] P. Gypens, et al., "Balanced Magnetic Logic Gates in a Kagome Spin Ice" Phys. Rev. Applied (2018)



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SP3 - Parallel Session 1

SP3.1.05

### Lieb-Mattis theorem and spin frustration in molecular nanomagnets

G. Kamieniarz, M. Antkowiak, W. Florek

**Text** Many quantum phenomena observed in molecular clusters are related to competing interactions between magnetic ions and can be accurately characterized in terms of spin models. On the basis of the fundamental Lieb-Mattis theorem (LTM) (see [1]), we discuss if bipartiteness of a spin system is opposite to frustration, what are its implications for the envisaged ground states and the classification of frustration as well as for the occurrence of the Kahn degenerated frustration.

Our approach relays on the graph theory and extensive numerical simulations. We consider a plethora of spin architectures with different number of sites, spin values and various configurations of interactions which occur in real materials. We demonstrate that the frustrated systems can be bipartite. Moreover, if an architecture of interactions break bipartiteness, the systems can still inherit the LMT implications and preserve the proper sequence of the ground states and a particular energy level ordering. This feature entails positions of the Kahn degenerated frustration points which correspond to the energy level crossings and enhance significantly the spin degeneracy dependent phenomena.

The findings are applicable to both the low-spin and the high-spin magnetic complexes, i.e. may help in tailoring emergent materials for storage and quantum information processing or magnetic field driven cooling.

[1] G. Kamieniarz et al., Phys. Rev. B92, 140411(R) (2015); Florek et al., Phys. Rev. B94, 224421 (2016).



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SP3 - Parallel Session 1

SP3.1.06

# High Spin Cycles: Topping the Spin Record for a Single Molecule verging on Quantum Criticality

J. Schnack

**Text** The cyclisation of a short chain into a ring provides fascinating scenarios in terms of transforming a finite array of spins into a quasi-infinite structure. If frustration is present, theory predicts interesting quantum critical points, where the ground state and thus low-temperature properties of a material change drastically upon even a small variation of appropriate external parameters.

Here we report a mixed 3d/4f cyclic coordination cluster that turns out to be very near or even at such a quantum critical point. It has a ground state spin of S=60, the largest ever observed for a molecule (120 times that of a single electron). [Fe<sub>10</sub>Gd<sub>10</sub>] forms a nano-torus with alternating gadolinium and iron ions with a nearest neighbour Fe-Gd coupling and a frustrating next-nearest neighbour Fe-Fe coupling. Such a spin arrangement corresponds to a cyclic delta or saw-tooth chain, which can exhibit unusual frustration effects. In the present case, the quantum critical point bears a 'flatland' of tens of thousands of energetically degenerate states between which transitions are possible at no energy costs with profound caloric consequences. Entropy-wise the energy flatland translates into a pointed summit overlooking the entropy landscape. Going downhill several target states can be reached depending on the applied physical procedure which offers new prospects for addressability.

A. Baniodeh et al., npj Journal Quantum Materials 3, 10 (2018)



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SP3 - Parallel Session 1

SP3.1.07

### A High-Temperature Quantum Spin Liquid with Atomic-Cluster Spins in 17-TaS<sub>2</sub>

D. Arcon

**Text** The Anderson's proposal of a resonating valence bond state [1] was put forward to explain the unusual magnetic properties of a triangular lattice of Ta atoms in the layered 17-TaS<sub>2</sub>. Compared to some recent candidates for quantum spin liquids (QSL), e.g., YbMgGaO<sub>4</sub> or  $\kappa$ -(ET)<sub>2</sub>Cu<sub>2</sub>(CN)<sub>3</sub>, 17-TaS<sub>2</sub> has a perfect triangular lattice geometry and a weaker spin-orbit coupling, offering a possibility for obtaining a unique insight into the competition between antagonistic QSL and Néel states.

Here we report [2] on QSL that appears to be realized by atomic-cluster spins on the triangular lattice of a low-temperature charge-density wave state of 1T-TaS<sub>2</sub>. In this system, nuclear quadrupole resonance and muon spin relaxation experiments reveal that the spins show gapless quantum spin liquid dynamics and no long range magnetic order down to 70 mK. Canonical T<sup>2</sup> power-law temperature dependence of the spin relaxation dynamics characteristic of a QSL is observed from 200 K to T<sub>f</sub> = 55 K. Below this temperature we observe a new gapless state with reduced density of spin excitations and high degree of local disorder (deduced from the decrease of stretching exponent *p* to

from the QSL. The QSL is at first robust against applying chemical pressure in doped samples before giving a way to superconducting state.

1. P. W. Anderson, Materials Research Bulletin 8, 153 (1973).

2. M. Klanjšek et al., Nature Physics 13, 1130 (2017).

~ 0.5) signify



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SP3 - Parallel Session 2

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SP3.2.01

### Elementary excitations and stability of the spin-stripe state in a quantum spin chain

M. Pregelj, A. Zorko, M. Gomilšek, D. Arčon, O. Zaharko, H. Luetkens, F. Coomer, H. Berger

**Text** Elementary excitations are a basic concept essential for the describe dynamical processes in condensed matter. Their variety is vast; ranging from established phonons and magnons to exotic magnetic monopoles [1] and Majorna fermions [2]. Yet, when order parameters intertwine, identification of elementary excitations is extremely difficult. For instance, the elusive excitations in high-temperature superconductors that propel fluctuating-charge-stripe, i.e., electronic nematic, phases [3] are still unidentified. Here we report on a new type of elementary excitations in a spin-stripe state, which occur as two perpendicular amplitude-modulated magnetic components with different modulation periods slide through each other. Exploring the frustrated zigzag spin-1/2 chain compound β-TeVO<sub>4</sub> [4,5] by muon-spin relaxation, neutron diffraction and dielectric spectroscopy, we find that the spin-orbit coupling introduces sizable anisotropic- and fourth-order-exchange interactions, which stabilize the spin-stripe phase and set the energy scale of underlying excitations. β-TeVO<sub>4</sub> offers a unique perspective on the stripe physics that avoid the problem of intertwining degrees of freedom, which hinders high-temperature superconductors.

- [1] C. Castelnovo et al., Nature 451, 42 (2008).
- [2] A. Banerjee et al., Nat. Mater. 15, 733 (2016).
- [3] R. M. Fernandes et al., Nature Phys. 10, 97 (2014).
- [4] M. Pregelj et al., Nature Commun. 6, 7255(2015).
- [5] M. Pregelj et al., Phys. Rev. B 94, 081114 (2016).



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SP3 - Parallel Session 2

SP3.2.02

### Spin dynamics of the classical spin nematic spinel LiGa0.95In0.05Cr4O8

G. J. Nilsen, R. Wawrzynczak, Y. Tanaka, M. D. Le, T. Guidi, Y. Okamoto, Z. Hiroi, M. Takigawa

**Text** The combination of magnetic frustration and strong spin-lattice coupling can lead to a wide variety of interesting magneto-structural behaviours. This is exemplified by the breathing pyrochlore spinels LiGaxIn1-xCr4O8, where both end-member compounds (x=0 and x=1) exhibit complex magneto-structural phase transitions on cooling. We have recently shown that replacing 5% Ga with In (x=0.05) suppresses this complexity, instead replacing it with a single transition to a possible collinear spin nematic state [1]. To examine the spin dynamics in this state, we have subsequently carried out an inelastic neutron scattering experiment on a powder sample. The high-temperature response is found to be quasi-elastic, with the characteristic energy displaying a power-law temperature dependence. Below the nematic transition, the spectrum is dominated by an inelastic mode, ascribed to local spin rotations lifted to finite energy by the magneto-elastic coupling. The appearance of similar modes in other lightly doped spinels raises the interesting possibility that nematic states may be more common than previously expected.

[1] R. Wawrzynczak et. al. PRL 119, (2017) 087201



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SP3 - Parallel Session 2

SP3.2.03

# Artificial spin systems as experimental simulators of frustrated magnets: from the fragmentation of magnetization to the six vertex model

N. Rougemaille, Y. Perrin, I.-A. Chioar, V.-D. Nguyen, M. Hehn, D. Lacour, F. Montaigne, A. Locatelli, T. O. Mentes, B. Santos Burgos, B. Canals

**Text** This talk aims at providing two examples of artificial spin systems in which the low-energy physics of two exotic Ising models was probed. The first one is related to the seminal six vertex model. More specifically, we show that a scan through the phase diagram of the six vertex model can be achieved experimentally [1]. In particular, the symmetric point of the square ice is recovered, and signatures of an algebraic Coulomb spin liquid are observed. Because of the experimental procedure used to reach this low-energy physics, quasi-particles are trapped in a disordered manifold, pointing to the need of thermal systems, but also emphasizing that these systems may be well suited for studying out-of-equilibrium relaxation of monopole-monopole pairs. The second example refers to a recent proposal, the fragmentation of magnetization [2], in an Ising kagome model. Here, we report the observation of this intriguing phenomenon, which corresponds to the splitting of the local degree of freedom into two channels, one ordering at low effective temperatures, in an antiferromagnetic all-in all-out fashion despite the ferromagnetic nature of the system, and the other, building a Coulomb-like low-energy manifold, inside which the magnetic equivalent of the Kirchhoff law at each node of the kagome lattice is fulfilled [3].

[1] Y. Perrin et al., Nature 540, 410 (2016).

- [2] M. E. Brooks-Bartlett et al., Phys. Rev. X 4, 011007 (2014).
- [3] B. Canals et al., Nat. Commun. 7, 11446 (2016).



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SP3.2.04

### Superspin-glass type behavior in single- and polycrystalline supercrystals

M. Smik, G. Wilbs, E. Kentzinger, U. Rücker, S. Mattauch, O. Petracic, T. Brückel

**Text** Magnetic nanoparticle (NP) assemblies form a novel type of artificial material which hold the promise to display properties that are not found in nature [1]. It is known that dense randomly closed packed arrangements behave like a canonical spin glass, and are therefore termed superspin glasses [2, 3]. Using a novel centrifuge assisted sedimentation technique [4], we have succeeded in fabricating large 3D NP arrangements composed of spherical Fe2<sub>2</sub>O<sub>3</sub> particles.

The size and size distribution of the individual NP were characterized using small angle x-ray scattering (SAXS).

The morphology of these crystals was examined using scanning electron microscopy (SEM), which showed that the growth of macrocrystals up to 300µm in size was possible. Their structure was then investigated using SEM and SAXS.

The magnetic behavior was characterized using magnetometry. The microscopic magnetic correlations between the NP were investigated using small angle neutron scattering. The systems exhibit characteristics of spin glasses, but show intriguing deviations from canonical spin glasses, specifically an inverted memory effect. This hints at a novel class of frustrated magnetic behavior.

[1] Disch et al., Nano Lett. 2011 11, pp. 1651-1656
[2] De Toro et al., Appl. Phys. Let. 102, 183104 (2013)
[3] S. Bedanta, O. Petracic, and W. Kleemann, Supermagnetism, Handbook of Magnetic Materials, Vol. 23, Ed. K.J.H. Buschow, Elsevier (2015)
[4] Wilbs et al., Materials Today: Proceedings 4, 2, pp. 146-153 (2017)



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SP3 - Parallel Session 2

SP3.2.05

#### A classification of frustration from topology

K. Roychowdhury, M. Lawler

**Text** We study the relationship between the physics to topology and frustration. Topological states of matter exhibit zero modes which are distinct from the ones arising from symmetry breaking. In other words, a zero mode must exist at the interface of two topologically distinct states. On the other hand, the accidental degeneracy of zero modes is a prominent aspect of frustrated systems as well. Taking cues from these two apparently different phenomena, we classify types of frustration using topology. We introduce an essential element in the formalism, the rigidity matrix, which is a non-Hermitian matrix and decides the topology of the zero modes

in a frustrated magnet. Further developments of the theory rely on combining the recent developments in our understanding of Maxwell constraint counting and generalizing the ten-fold way classification of Hermitian matrices to non-Hermitian matrices. The result is a three-fold way classification for each Maxwell counting index including both the isostatic and non-isostatic magnetic systems. We illustrate the classification by demonstrating the existence of a new vortex-like invariant for real rigidity matrices using random matrices and models of KHAF. Surprisingly in the latter, we discover topological properties of kagome coplanar states. So by classifying all the rigidity matrices, we answer the question of the origin of frustration in the form of accidental degeneracy in a wide class of frustrated magnets by linking it to topological invariants.



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SP3 - Parallel Session 2

SP3.2.06

#### Collective magnetism in an artificial 2D XY spin system

N. Leo, S. Holenstein, D. Schildknecht, O. Sendetskyi, P. Derlet, H. Luetkens, S. Lee, V. Scagnoli, K. Hofhuis, D. Lançon, J. Mardegan, L. Heyderman

**Text** Low-dimensional magnetic systems with continuous spin degrees of freedom, so-called XY moments, exhibit a rich spectrum of thermal behavior due to the strong competition between fluctuations and correlations [1]. In this work, we investigate the magnetic order in dipolar-coupled artificial XY spin systems manufactured from thermally-active nanoscale magnetic disks arranged on a square lattice.

Using low-energy muon spin relaxation [2], we observe correlated dynamics at the critical temperature and the emergence of static magnetism for strongly-coupled nanomagnets. Further information on the spatial correlations of the artificial XY spin systems are obtained by soft x-ray resonant scattering [3], and we find clear indications of a unit-cell doubling, supporting the formation of striped magnetic order below the critical temperature.

With these results [4] we demonstrate emergent correlations in thermally-active artificial spin systems featuring continuous spin degrees of freedom, opening new possibilities to study the magnetic behavior of two-dimensional analogues of spin ices.

References:

- [1] S. Prakash & C.L. Henley, Phys. Rev. B 42 6574-6589 (1990)
- [2] L. Anghinolfi et al., Nat. Comm. 6 8278 (2015)
- [3] O. Sendetskyi et al., Phys. Rev. B 93 224413 (2016)
- [4] N. Leo et al., under review.



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SP3 - Parallel Session 2

SP3.2.07

#### Interaction radius influence on phase transition in artificial kagome spin ice

P. Andriushchenko, K. Nefedev

**Text** Two types of samples (square and hexagonal forms) which are consisting of various numbers of particles formed in artificial kagome spin ice arrays with the different radius of dipole interaction were simulated by multicanonical Wang-Landau method. Models were considered with long-range (LR, when each dipole interacts with each other) and short-range (SR, when each dipole interacts only with the nearest neighbors) dipole-dipole interaction. The temperature dependence of the heat capacity in LR interaction models shows anomalous character, there are two temperature peaks, i.e., critical behavior of the system of dipoles in the region of critical temperature significantly changes. The first peak with increasing number of particles increases and the second peak reduces. The heat capacity peak of models with SR interaction with increasing number of particles is reduced. In models with SR interaction, the phase transition is absent, while models with LR interactions show the phase transition. Thus, the phase transition is provided by distant neighbors, and not by short-range ones. An original order parameter allowing to separate the ordered phase from the disorder for LR model is proposed. The order parameter is a percolation cluster formed by vortices of hexagons in the lattice.



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SP3 - Parallel Session 2

SP3.2.08

### Exotic phenomena in the new frustrated two-leg ladder Li2Cu2O(SO4)2

O. Vaccarelli, A. Honecker, A. Saul, P. Giura, B. Fåk, G. Rousse, G. Radtke

**Text** Despite decades of theoretical work devoted to the study of frustrated spin ladders (see, e.g., [1] and references therein), real material realizations of such systems still remain limited. In this work, we investigate the magnetic properties of the new compound Li2O(CuSO4)2 [2], which appears as a rare realization of a frustrated two-leg spin ladder in its high-temperature tetragonal structure [3]. Through a combined experimental and theoretical approach, we demonstrate that most of the magnetic frustration is removed by a structural transition, occurring at about 125K, and leading to a strong magnetic dimerization of the Cu ions [4]. Furthermore, we present the first detailed investigation of the low-temperature magnetic excitations of Li2Cu2O(SO4)2 combining magnetic susceptibility, infrared spectroscopy and inelastic neutron scattering measurements. All these observations are qualitatively explained by higher-order perturbation calculations carried out on the basis of the dimerized geometry derived from previous first principle calculations.

- [1] T. Vekua and A. Honecker, Phys. Rev. B 73, 214427 (2006).
- [2] M. Sun, et al., Chem. Mat. 27, 3077 (2015).
- [3] G. Rousse, et al., Phys Rev B 95, 144103 (2017).
- [4] O. Vaccarelli, et al., Phys. Rev. B 96, 180406(R) (2017).



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SP3 - Parallel Session 3

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SP3.3.01

### Imaging the magnetic stray field of an artificial chiral spin ice system

M. Wyss, D. Vasyukov, S. Gliga, L. Ceccarelli, G. Romagnoli, R. L. Stamps, M. Poggio

**Text** We use a scanning nanometer-scale superconducting quantum interference device (nanoSQUID) [1] to image the magnetic stray field of a chiral artificial spin ice system in a series of in- and out-of-plane applied magnetic fields at 4.2 K. The measured stray field maps are largely consistent with micromagnetic simulations of the expected patterns. For large in-plane applied fields, however, we observe a distorted pattern, indicating a bending of the local magnetization at the edge of each island and therefore a breakdown of the single-spin approximation [2]. At low applied fields, complex stray field patterns emerge likely due to the breakup of the magnetization into multiple domains. This first application of scanning nanoSQUID to spin ice, which can achieve lateral resolutions and sensitivities better than 50 nm and 10 nT//Hz, respectively, is particularly promising for these systems. Unlike magnetic force microscopy, which measures magnetic force and from which precise stray fields values are often difficult to extract, scanning nanoSQUID produces quantitative maps of magnetic flux. These flux distributions provide insight into the behavior of the system and therefore provide a better picture of its state.

[1] D. Vasyukov, Y. Anahory, L. Embon, D. Halbertal, J. Cuppens, L. Neeman, A. Finkler, Y. Segev, Y. Myasoedov, M. L. Rappaport, M. E. Huber, and E. Zeldov, Nat Nano 8, 639 (2013)

[2] S. Gliga, A. Kákay, L. J. Heyderman, R. Hertel, and O. G. Heinonen, PRB 92, 060413(R) (2015)



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SP3 - Parallel Session 3

SP3.3.02

#### Interaction Induced Special Temperatures in Frustrated Magnets and Gases

L. Bovo, M. Twengström, O. Petrenko, T. Fennell, M. Gingras, S. Bramwell, P. Henelius

**Text** The magnetic bulk susceptibility can be used to classify magnets as ferromagnets (chi T/C >1) or antiferromagnets (chi T/C <1). In this study we identify a new class of ``inverting" magnets that exhibit a maximum in

chi T/C as a function of temperature. In strong analogy with van der Waals theory of classical gases we identify the

peak temperature with a magnetic Joule temperature, where the system is quasi-ideal, dU/dM=0, and the onset of

antiferromagnetic correlations. In addition, we find a magnetic Boyle temperature, where chi T/C=1, and the incipient

ferromagnet turns to an antiferromagnet at low temperature. We provide a phenomenological model of the susceptibility which reveals the mechanism that induces the special temperatures and elevates the effects of minute

frustrated exchange interactions to surprisingly high temperature. By explicitly decomposing the dipolar Hamiltonian

we demonstrate that these special temperatures and eventual antiferromagnetic ordering are caused by the quadrupolar corrections to a monopolar (dumbbell) Hamiltonian. Our study establishes chi T/C as a direct measure of

interaction parameters which are otherwise difficult to access experimentally. We find that the spin ice materials

Dy/Ho2Ti2O7, Kapellasite, several spin glass materials and the Ising and Heisenberg models on the square lattice

belong to this class of magnets.



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SP3 - Parallel Session 3

SP3.3.04

### Alloy disorder and fluctuating magnetic moments in the Earth's core

V. Drchal, J. Kudrnovsky, D. Wagenknecht, I. Turek

**Text** The electronic and thermal transport properties of the Earth's core are crucial for many geophysical models such as the geodynamo model of the Earth's magnetic field. We show by first-principles modeling and methods of statistical physics that the spin disorder, not considered in previous studies, gives an essential contribution to the electrical resistivity at the Earth's core conditions. The origin of this spin-disorder resistivity (SDR) consists in the existence of fluctuating local moments that are stabilized at high temperatures by the magnetic entropy even at pressures at which the ground state of iron is non-magnetic. It turns out that the contributions of various scattering mechanisms (alloy disorder, phonon scattering, spin disorder, and electron-electron correlations) are comparable, but not additive [1]. Here we report results for iron and iron-rich alloys (Fe-O, Fe-Si, Fe-S) that can be present in the Earth's core. Special attention is paid to alloys with two magnetic elements (Fe-Ni).

Reference

[1] V. Drchal, J. KudrnovskÃ<sup>1</sup>/<sub>2</sub>, D. Wagenknecht, I. Turek, and S. Khmelevskyi: Phys. Rev. B 96, 024432 (2017).



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#### Critical behaviour and evidence of low-energy orbital excitations in $\gamma$ -Li<sub>2</sub>FeSiO<sub>4</sub>

R. Klingeler, C. Neef, W. Hergett, J. Werner, F. Billert, M. Jonak, S. Gu, C. Koo, M. Abdel-Hafiez, H. Wadepohl, M. Haverkoort

**Text** Large Li<sub>2</sub>FeSiO<sub>4</sub> single crystals featuring the high temperature Pmnb-phase ( $\gamma$  -Li<sub>2</sub>FeSiO<sub>4</sub>) were grown for the first time by the optical floating zone method under high Ar-pressure and the single crystal structure was solved. We report specific heat, thermal expansion, magnetostriction, and high-frequency electron spin resonance (HF-ESR) studies up to 16 T and magnetization data up to 60 T. The chain-like magnetic substructures of tetrahedrally coordinated  $S = 2 \text{ Fe}^{2+}$ -moments evolve long-range antiferromagnetic order at  $T_N = 17 \text{ K}$ . Long range order is suppressed by B = 16 T to lowest temperatures where a possibly quantum critical transition to high-field antiferromagnetic phases is observed. The uniaxial pressure dependencies are derived from thermal expansion data and the magnon modes detected by HF-ESR imply an anisotropy gap of 125 GHz. Remarkably, sizable magnetostriction, anomalous entropy changes and strong temperature effects on the ESR resonance features imply magneto-structural effects and/or low energy excitations up to around 150 K. We discuss the results in terms of short range liquid-like orbital order and low-energy orbital excitations within the low-lying  $e_g$ -orbitals.



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# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 3

SP3.3.06

### Experimental investigation of kagomé artificial colloidal ice

A. Le Cunuder, A. Ortiz-Ambriz, N. Rougemaille, I. Frérot, P. Tierno

**Text** We study magnetic frustration through an experimental system developed in the group: we reproduce the analog of a spin-ice system by filling sculptured double-well traps with repulsively interacting magnetic colloids. Each double-well can trap one particle. The traps are arranged in a square or in an hexagonal lattice realizing an artificial ice. Colloidal Artificial Spin Ice can be described in the same way as magnetic spin ice by defining a spin lying along the trap and pointing towards the particle.

Within this framework, an important issue is to explore the low temperature physics of a kagome system. More precisely, we are investigating the phase diagram of our colloidal system, in the quest for the ground-state manifold. Indeed, Brownian simulations suggest that our colloidal system cannot be mapped exactly onto a dipolar spin ice system, leading to a new, still unexplored, phenomenology. Starting from a random configuration, we study experimentally how the system self-organizes when a magnetic field is applied, and if some order spontaneously builds up. Analyzing the structure of correlations, we aim at describing our system by an effective thermodynamics.



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# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 3

SP3.3.07

### Dynamical magnetic properties of interacting ultra-small nanoparticle systems

R. Mathieu, M. Hudl Waltin, P. Nordblad, M. Vasilakaki, G. Margaris, K. N. Trohidou, N. Yaacoub, M. Bellusci, F. Padella, D. Fiorani, D. Peddis

**Text** The magnetic interaction between nanoparticles brings forth collective states such as superspin glass states. The dynamical magnetic properties of superspin glasses are qualitatively similar to those of atomic spin glasses. That includes a (super)spin glass phase transition, and dynamical features such as aging, memory, and rejuvenation phenomena[1]. We have here investigated the superspin glass state ( $T_g \sim 46$  K) of a dense assembly of 2 nm MnFe<sub>2</sub>O<sub>4</sub> nanoparticles by magnetometry, recording the zero-field-cooled (ZFC), thermo- and isothermal remanent (TRM and IRM) magnetization after specific protocols [1]. We find that, in spite of the significant disorder affecting inter- and intraparticle interactions, the glassy state displays the same dynamical features and superposition as atomic spin glasses. Monte Carlo simulations considering a mesoscopic model of the nanoparticle system [2] reproduce well the experimental findings. The effect of the superspin dimensionality is discussed[3].

The Swedish Research council (VR) and the Wenner-Gren Foundations are thanked for financial support.

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[3] M. S. Andersson, J. A. De Toro, S. S. Lee, P. S. Normile, P. Nordblad, and R. Mathieu, Phys. Rev. B **93**, 054407 (2016).



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# SP 3 Frustrated and disordered magnetism

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**SP3 - Parallel Session 4** 

SP3 - Parallel Session 4

SP3.4.01

### Exchange-anisotropy and frustration in epitaxial thin films of SrMn<sub>0.5</sub>Ir<sub>0.5</sub>O<sub>3</sub>

S. Dasgupta, P. Komissinskiy, I. Radulov, L. Alff

**Text** We present our studies on highly crystalline epitaxial thin films of a 3d-5d based perovskite SrMn<sub>0.5</sub>Ir<sub>0.5</sub>O<sub>3</sub> (SMIO) grown by pulsed laser deposition on single-crystalline SrTiO<sub>3</sub> (001) substrates. Magnetic measurements of the SMIO films reveal  $T_c = 110$  K with a magnetic saturation of 0.4  $\mu$ B /f.u. at 5 K. A low difference in the ionic radii and similar formal valence of Mn<sup>4+</sup> and Ir<sup>4+</sup> in SMIO initiate exchange-anisotropy. An exponential relaxation behaviour with time-dependence confirms frustration via spin-glass. Temperature dependent electrical resistivity measurement indicates that SMIO is a narrow band-gap insulator, which in addition to the observed exchange-anisotropy gives rise to magneto-resistance in the system.



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# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 4

SP3.4.02

### Magnetic order and single-ion anisotropy in frustrated magnet - Tb3Ga5O12

R. Wawrzynczak, B. Tomasello, P. Manuel, D. Khalyavin, M. D. Le, T. Guidi, A. Cervellino, T. Ziman, G. Nilsen, T. Fennell

**Text** Terbium gallium garnet (Tb3Ga5O12, TGG), hosts two inter-penetrating half-garnet lattices (3D twisted arrays of corner-sharing triangles) forming geometrically frustrated Tb3+ sublattice. It is the member of the rare-earth garnet family, known for their numerous applications in optoelectronic devices (e.g. lasing media, Faraday rotators), characterized by one of the highest-known Vedet constants (efficiency of Faraday rotation in the material). TGG has also exhibited various magnetoelastic phenomena i.e. paramagnetic acoustic Faraday rotation and thermal Hall effect, becoming the prototypical system for the latter one.

Neutron diffraction studies of the ordered magnetic phase have shown its strongly anisotropic structure -Tb3+ moments lying parallel and anti-parallel to the three main crystallographic directions of the cubic unit cell and the ordered moment having around a third of the free ion value. Significant ratio between Tn=240 mK and |Tcw|=7.98 K suggests the presence of strongly correlated, but disordered phase in between, confirmed by presence of diffuse scattering persistent beyond the |Tcw|. Inelastic neutron scattering has provided the ground-state splittings and allowed for refinement of crystal field parameters consistent with the bulk measurements results in non-correlated regime. It has also shown the complex structure of the groundstate quasi-doublet (dE~0.22 meV), with higher level giving rise to multiple exciton branches following a robust T-dependence.



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# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 4

SP3.4.03

#### The AC susceptibility of nanocomposite rare earth titanates

A. Rinkevich, D. Perov, O. Nemytova

**Text** The AC susceptibility of R2Ti2O7 type rare earth nanocomposite titanates, where R = Dy, Nd, Sm, Er, Gd, Pr, has been studied within the frequency range from 10 Hz to 10 kHz. The nanocomposites are the opal matrices containing titanate particles sized from 5 to 60 nm which are placed into the inter-spherical areas. The measurements have been performed at the temperature range from 2 to 30 K in the magnetic fields up to 30 kOe. The Argand diagrams have been constructed and approximation of temperature dependences within the framework of Cole-Cole model has been performed. It has been established that the Argand diagrams for nanocomposite titanates can be constructed upon the change of susceptibility not only with the change of frequency but also with the change of magnetic field. The values of adiabatic and isothermal susceptibilities have been obtained and their dependencies on temperature and magnetic field have been analyzed. As a rule, the real part of susceptibility of nanocomposites decreases upon the increasing of magnetic field. The imaginary part of susceptibility decreases sharply upon the increasing of temperature. For nanocomposites with Gd2Ti2O7 and Er2Ti2O7 particles, there is a maximum on the field dependence of imaginary part of susceptibility.



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# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 4

SP3.4.04

#### Anomalous excitation in frustrated magnets under the external magnetic fields

S. Miyahara

**Text** A helical magnet is stabilized due to frustration of the magnetic interactions and/or Dzyaloshinsky-Moriya interaction and known as a typical magnetoelectric multiferroics where there is a strong coupling between magnetization and electric polarization. We investigate magnetoelectric couplings between an electric polarization, spin and orbital in a multiorbital Hubbard model on a distorted lattice. We microscopically clarify the origin and nature of the magnetoelectric couplings [1]. Such couplings induce anomalous effects even in dynamical processes. One of the examples is an electro active spin wave excitation (so called electromagnon excitation) [2]. Such spin wave may show novel features as spin-wave spin current. We clarify the dynamics of the spin-wave spin current in a helical magnet. When the external magnetic field is applied in the helical magnets, the conical spin structure is realized. We show that the spin-wave spin current in the conical magnets show the non-reciprocal property, i.e., the way of the propagation of the spin wave depends on the direction of the propagation. Such nonreciprocal property can be tuned not only by the external magnetic fields but also the external electric fields due to the magnetoelectric coupling.We mention possible anomalous excitation in frustarted magnets.

[1] S. Miyahara and N. Furukawa, Phys. Rev. B 93, 01445 (2016)

[2] H. Katsura, A.V. Balatsky, and N. Nagaosa, Phys. Rev. Lett. 98, 027203 (2007).



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SP3 - Parallel Session 4

SP3.4.05

### Magnetisation process in the rare earth tetraborides, NdB4 and HoB4

O. Petrenko, D. Brunt, G. Balakrishnan, D. Mayoh, M. Lees, D. Gorbunov, N. Qureshi

Text A field-induced magnetisation process in the frustrated antiferromagnets is often much richer compared to the materials without competing interactions. The applied field tends to stabilise unusual spin configurations which frequently results in the appearance of magnetisation plateaux. Here we report a study into the field-induced magnetisation of the two frustrated rare earth tetraborides, HoB4 and NdB4. NdB4 shows a fractional magnetisation plateau occurring at M/Msat≈1/5 before saturating in a field of 33 kOe. On cooling down to 0.5 K the temperature dependent susceptibility of NdB4 shows an unconventional transition where the system returns to the zero field antiferromagnetic state from a higher-temperature ferrimagnetic state. We are able to reconstruct the magnetic phase diagram of NdB4 from the magnetisation, susceptibility and resistivity measurements for both H//c and H ⊥ c. For HoB4, the interesting behaviour is found at the lowest temperature of 0.5 K, where the field dependent magnetisation demonstrates a new fractional 1/2 magnetisation plateau. Further insight into the relations between the exchange interactions and single ion effects is gained through high-field magnetisation measurements in both HoB4 and NdB4.



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# SP 3 Frustrated and disordered magnetism

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SP3 - Parallel Session 4

SP3.4.06

#### Topological classification of magnetic plateaux in spin-1/2 Heisenberg multimer chains

I. Maruyama, S. Miyahara

**Text** Spin-1/2 Heisenberg multimer chains as a generalization of orthogonal-dimer chain have magnetic plateaux and exact ground states in the case of symmetric frustration.[1] To classify magnetic plateau by using topological invariant, we adopt fractionally quantized Berry phase, which has been demonstrated in the non-interacting tight-binding models[2]. The Berry phase for this spin model is defined by local spin twists in the spin xy plane. The spin twists are introduced into inner-multimer Heisenberg coupling terms of a local multimer (N-mer). Fractional quantization of the Berry phase comes from N-fold rotational symmetry of N-mer in the same way as Ref.[2]. The Z\_N Berry phases are calculated by numerical integration with using the ground states obtained by the Lanczos method. As a result, we obtained fractional quantization of Z\_2 and Z\_4 Berry phases in dimer (N=2) and tetramer (N=4) chains respectively. Especially, in the tetramer case, six magnetic plateaux appeared in the phase diagram of positive magnetic field are classified by four kinds of Berry phases, correspond to 0, 90, 180, 270 degrees. Meaning of these angles can be understood by magnetization in the decoupled N-mer limit. As a merit of quantized invariants, topological identification works well even in a generic model whose ground state cannot be rigorously obtained. [1] A.Honecker, et.al., Eur.Phys.J.B. 15, 227(2000), [2] Y.Hatsugai, IM, Eur.Phys.Lett. 95, 20003 (2011)



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SP3 - Parallel Session 4

SP3.4.07

# Magnetic ordering and open volume defects – phase transitions in ion irradiated $Fe_{\rm 60}AI_{\rm 40}$ thin films

M. O. Liedke, J. Ehrler, R. Bali, M. Butterling, E. Hirschmann, A. Wagner

**Text** Fe<sub>60</sub>Al<sub>40</sub> exhibits the so-called *disorder induced ferromagnetism*, where *anti-site disorder* (ASD) promotes ferromagnetic A2-phase (disordered) over paramagnetic B2-phase (ordered). Both phases can be - in a controllable fashion - driven by ion irradiation or annealing, respectively. The main physical origin correlates strongly with ASD [R. Bali, et al., *Nano Lett.* **14**, 435 (2014)]. Nevertheless, the concentration and size of open volume defects can be of crucial importance in determining the kinetics of the reordering processes. To unravel the influence of vacancy clusters, three different initial order states have been investigated: (i) as-sputtered, (ii) as-grown irradiated with Ne<sup>+</sup> and (iii) B2 ordered films, obtained via 773 K annealing and Ne-irradiated. Open volume defects in the treated samples were investigated with Doppler broadening and positron annihilation lifetime spectroscopy. Furthermore, since the reordering directly affects M<sub>s</sub>, the extent of the diffusion process can be traced via magnetometry at slightly elevated temperature of 400 K. We show that immobile large vacancy clusters are dominant in the as-grown films; these complexes present only in the as-sputtered film possess a high thermal activation barrier and hinder ordering. Ion irradiation breaks down these pinning defects strongly accelerating thermal diffusion and reordering. These results provide insights into thermal reordering processes in binary alloys, and the consequent effect on magnetic behavior.



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SP3 - Parallel Session 4

SP3.4.08

# Observation of a 4-spin Plaquette Singlet State in the Shastry-Sutherland compound SrCu2(BO3)2

M. Zayed, C. Ruegg, J. Larrea J, A. M. Laeuchli, C. Panagopoulos, S. S. Saxena, M. Ellerby, D. F. McMorrow, T. Straessle, S. Klotz, G. Hamel, R. Sadykov, V. Pomjakushin, M. Boehm, M. Jiménez–Ruiz, A. Schneidewind, E. Pomjakushina, M. Stingaciu, K. Conder, H. M. Ronnow

**Text** The study of interacting spin systems is of fundamental importance for modern condensed matter physics. On frustrated lattices, magnetic exchange interactions cannot be simultaneously satisfied, and often give rise to competing exotic ground state. The frustrated 2D Shastry-Sutherland lattice realized by SrCu2(BO3)2 is an important test to our understanding of quantum magnetism. It was constructed to have an exactly solvable 2-spin dimer singlet ground state within a certain range of exchange parameters and frustration. While the exact dimer state and the antiferromagnetic order at both ends of the phase diagram are well-known, the ground state and spin correlations in the intermediate frustration range have been widely debated. We report here the first experimental identification of the conjectured 4-spin plaquette singlet intermediate phase in SrCu2(BO3)2. It is observed by inelastic neutron scattering after pressure tuning at 21.5 kbar. This gapped singlet state leads to a transition to an ordered Neel state above 40 kbar, which may represent a deconfined quantum critical point.

M. E. Zayed et al., 4-spin Plaquette Singlet State in the Shastry–Sutherland compound SrCu2(BO3)2, Nature Physics 13, 962–966 (2017)